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Subpicosecond Luminescence Studies of Carrier Dynamics in Nitride Semiconductors Grown Homoepitaxially By MBE On GaN Templates

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ABSTRACT

A new technique is presented that employs luminescence downconversion using an ultrashort gating pulse to enable the characterization of UV light emission from III-nitride semiconductors with subpicosecond temporal resolution. This technique also allows one to measure PL rise times and fast components of multiple decays in the subsequent time evolution of the PL intensity. Comparison of luminescence emission intensity and lifetime in GaN and AlGaIn with ~0.1 Al content grown homoepitaxially on GaN templates with the same quantities measured in heteroepitaxial layers grown on sapphire indicate significant improvement in the homoepitaxial layers due to reduction in dislocation density. Fast (<15 ps) initial decays in the AlGaIn are attributed to localization associated with alloy fluctuations and subsequent recombination through gap states.

INTRODUCTION

One of the most important issues in the development of high quality III-nitride semiconductors is the lack of native substrates for epitaxial growth. Most devices are fabricated on either sapphire or SiC substrates, with the resulting material quality often dictated by the control of threading dislocations produced by the lattice mismatch between the III-nitride epilayers and these substrates. These threading dislocations lead to enhancement of nonradiative recombination that reduces radiative efficiency in ultraviolet light emitters [1], as well as degradation of the reliability of high sensitivity avalanche photodiodes [2]. Moreover, carrier localization associated with alloy fluctuations in the AlGaIn alloys required for operation in the important 280 nm to 340 nm spectral region can also lead to nonradiative recombination in UV emitters and reduced out of band rejection associated with broadening of the absorption edge in detectors [3-5]. Time-resolved photoluminescence (TRPL) provides information about radiative lifetimes and nonradiative recombination mechanisms in III-nitride semiconductors crucial to the design of UV light emitters and detectors. Moreover, it has been shown that reduction of dislocations in GaN leads to both longer PL [6-7] and longer carrier lifetimes [8]. These results imply that

homoepitaxial growth of III-nitride materials on low defect density GaN templates should lead to improved radiative efficiency and concomitant longer PL lifetimes at room temperature, and it is therefore important to evaluate the extent to which this is the case in both GaN and AlGaIn, for which it has been suggested that alloy fluctuations lead to ultrafast carrier localization [3]. Such studies have been hampered to date by the temporal resolution (~ 10 ps or larger) of current UV optoelectronic spectroscopic techniques involving streak cameras and time-correlated photon counting [4-7].

In this paper we present a new technique that employs luminescence downconversion using an ultrashort gating pulse to enable the characterization of UV light emission from III-nitride semiconductors with subpicosecond temporal resolution. This technique also allows one to measure PL rise times and fast components of multiple decays in the subsequent time evolution of the PL intensity. Comparison of luminescence emission intensity and lifetime in GaN and AlGaIn with ~ 0.1 Al content grown homoepitaxially on GaN templates with the same quantities measured in heteroepitaxial layers grown on sapphire indicate significant improvement in the homoepitaxial layers due to reduction in dislocation density. Observation of fast (< 15 ps) initial decays in the AlGaIn irrespective of substrate suggests that localization associated with alloy fluctuations and subsequent recombination through gap states is the dominant decay mechanism in this material.

EXPERIMENTAL METHOD

The samples for the optical studies were grown by molecular beam epitaxy using standard techniques on either c-plane sapphire substrates or thick (~ 5 -10mm) GaN templates deposited by hydride vapor phase epitaxy (HVPE) on sapphire substrates. In the absence of bulk substrates the use of thick templates is employed because threading dislocations propagating from the GaN-sapphire interface tend to annihilate with increasing film thickness, leading to improved optical properties of the GaN [6]. The dislocation density for these thin GaN templates was estimated to be in the mid- $10^8/\text{cm}^2$ range. In contrast, typical dislocation densities in thin GaN epilayers deposited by MBE directly on sapphire are greater than $10^9/\text{cm}^2$.

The time-resolved experiments were performed using a 250 kHz Ti:sapphire regenerative amplifier-pumped optical parametric amplifier that produces ultrashort pulses frequency doubled to obtain a source of UV pulses tunable between 225 nm and 375 nm. Photoluminescence (PL) was measured with subpicosecond resolution using a novel PL downconversion technique that provides one to two orders of magnitude better temporal resolution than standard ultraviolet time-resolved PL measurement techniques such as time-correlated photon counting or use of a streak camera and spectrometer [4-7]. In a downconversion experiment, the PL created by the radiative recombination of electron-hole pairs excited by an ultrafast ultraviolet pulse is time-resolved through gating of the PL in a nonlinear crystal with a synchronized ~ 800 nm pulse derived from the same source, the regenerative amplifier. Since the nonlinear process,

downconversion, is nonresonant, it only occurs within the temporal slice of the luminescence coincident in time with the gating pulse. In this way, the time dependence of the PL can be mapped out with subpicosecond resolution by varying the timing of the gating pulse with respect to that of the excitation pulse using a mechanical delay line. While this experiment is quite similar to PL upconversion studies performed on infrared emitting materials [9], the lack of efficient detectors in the deep UV for detection of the upconverted UV PL dictates that downconversion of this PL to the visible is required.

RESULTS AND DISCUSSION

The temporal resolution of our apparatus is demonstrated in fig. 1, which shows the rise time of the spectrally integrated bandedge PL from a GaN epilayer on an HVPE template photoexcited by a pump pulse centered at 348 nm. The system response, obtained by downconversion of pump light scattered from a frosted glass, is illustrated in the cross-correlation trace, which shows a full width at half maximum limited by the dispersion in the collection optics to ~ 250 fs. The PL intensity increases abruptly at first, but then transitions to a slower rise that reaches a plateau at ~ 1.5 ps. The fact that the rise time of the data is both significantly slower than the time integral of the cross-correlation and possesses multiple time scales indicates that the rise of the PL is temporally resolved for the first time. In this case the PL rise time contains information primarily about hot carrier thermalization and energy relaxation through carrier-LO phonon and carrier-carrier scattering.

Another important aspect of this temporal resolution is that it allows one to resolve fast components of multiple decays in the subsequent time evolution of the PL intensity that may

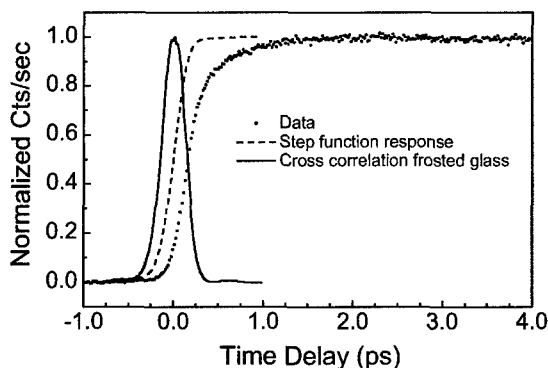


Fig. 1. Temporal resolution of the luminescence downconversion technique. The data provide a measure of the rise time of the spectrally integrated bandedge photoluminescence from GaN for excitation at 348 nm. The system resolution is obtained by downconversion of the pump pulse scattered from frosted glass.

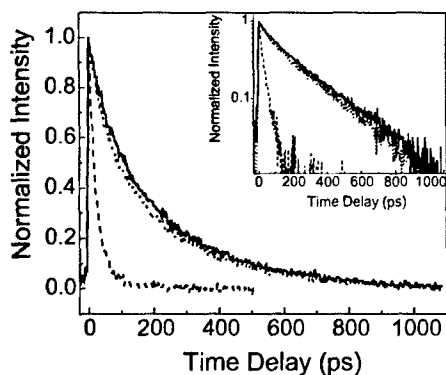


Fig. 2. Downconverted photoluminescence signal as a function of time delay for 364 nm excitation of GaN on HVPE GaN template (solid line), GaN on sapphire (dashed line), and GaN template (dotted line). Inset: data on log scale.

provide information about the effect of ultrafast trapping on radiative recombination. Figure 2 shows a comparison of the room temperature PL decays for bandedge (364 nm) excitation of GaN grown homoepitaxially on a thick, low defect density HVPE GaN template, and GaN grown heteroepitaxially on sapphire. The excitation density in these samples was in the mid- 10^{18} - $10^{19}/\text{cm}^3$ range. The linear plot shows that the PL lifetime in the homoepitaxially grown sample is significantly longer than that in its heteroepitaxially grown counterpart, and about the same as that in the GaN template itself. This result is consistent with the fact that the CW PL is nearly 20 times brighter for the homoepitaxial sample than for the heteroepitaxial one, while the linewidth is ~ 6 meV narrower (32 meV for GaN on template versus 38 meV for GaN on sapphire). The semilog plot in the inset indicates that while the GaN on sapphire data may be characterized primarily by a single exponential decay of ~ 20 ps, the GaN on template data possess both a fast (~ 65 ps) and a more dominant slow (~ 300 ps) decay. The fast decay in both samples may be associated with trapping and subsequent nonradiative recombination at defect-related states in the gap, with the longer decays in the homoepitaxial sample indicative of a lower defect density and saturation of these deep states. These results are consistent with ones obtained for high quality and low quality MOCVD-grown GaN on sapphire in which the dislocation densities were measured to be close to what we expect for our samples [7]. The PL decay in the high quality undoped material possessed time constants of 50 ps and 250 ps for a dislocation density of $4 \times 10^8/\text{cm}^2$, while that in the low quality undoped sample was characterized by a single 30 ps decay time for a dislocation density of $2 \times 10^9/\text{cm}^2$. Moreover, the fact that PL decay times of 530 ps for a 63 mm-thick HVPE template [6] and 860 ps for an 80mm-thick quasi-

substrate [10] suggests that even better results should be obtained for growth on thicker templates with lower defect densities.

Figure 3 shows a comparison of room temperature PL decays in AlGa_{0.1}N with ~0.1 Al content for 330 nm excitation. The PL decays for both AlGa_{0.1}N samples possess both a slow and a fast decay. The fast decay in both samples is ~13 ps, while the slow decay is longer (~61 ps) in the AlGa_{0.1}N on template than for the sample on sapphire (~39 ps), but still shorter than that for the GaN template (~83 ps). The fast PL decay, which has not been observed before due to lack of

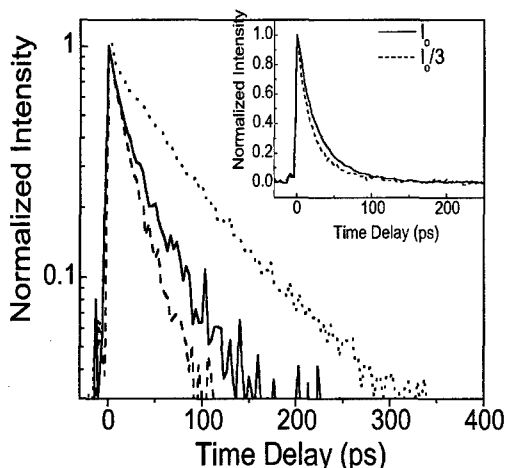


Fig. 3. Downconverted photoluminescence signal as a function of time delay for 330 nm excitation of Al_{0.1}Ga_{0.9}N on HVPE GaN template (solid line), Al_{0.1}Ga_{0.9}N on sapphire (dashed line), and GaN template (dotted line). Inset: Intensity dependence of PL decays in Al_{0.1}Ga_{0.9}N on sapphire.

temporal resolution [5] and occurs regardless of template/substrate, may be due to localization in shallow levels and subsequent nonradiative recombination associated with alloy fluctuations, with the decays becoming faster with decreasing intensity in the sample on sapphire (inset), as observed in time-resolved reflectivity experiments [3]. Low temperature CW PL measurements show an alloy broadening of ~23 meV in the PL linewidth when moving from GaN to AlGa_{0.1}N with 0.1 Al content [11]. These results are consistent with the measured room temperature linewidths of 55 meV and 61 meV for our homo- and heteroepitaxial AlGa_{0.1}N, respectively, when viewed in the context of our GaN PL linewidths reported earlier. The discrepancy in light emission dynamics between the homoepitaxial AlGa_{0.1}N and the GaN template may also be due to the combination of carrier localization and nonradiative recombination associated with alloy fluctuations in the AlGa_{0.1}N. The longer decays in these AlGa_{0.1}N samples may be associated with

trapping and nonradiative recombination through deep states related to dislocations and other defects, as in GaN. Viewed in this way, the time-resolved PL data suggest that homoepitaxial growth of AlGaIn on GaN templates leads to improved material quality, even though the TRPL on the GaN template employed in this case indicates that this template is of poorer structural quality than the one employed for the GaN homoepitaxial growth.

CONCLUSION

In conclusion, we have demonstrated a unique luminescence downconversion technique for the measurement of UV PL from III-nitride semiconductors with subpicosecond temporal resolution. The ability of this technique to resolve both PL rise times and fast components of multiple decays in the subsequent time evolution of the PL intensity enables the study of carrier relaxation and the effect of ultrafast trapping on radiative recombination. Comparison of luminescence emission intensity and carrier lifetime in GaN and AlGaIn with ~0.1 Al content grown homoepitaxially on GaN templates with the same quantities measured in heteroepitaxial layers grown on sapphire indicate significant improvement in the homoepitaxial layers due to reduction in defect density. Fast (<15 ps) initial decays in the AlGaIn are attributed to localization associated with alloy fluctuations and subsequent recombination through gap states.

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